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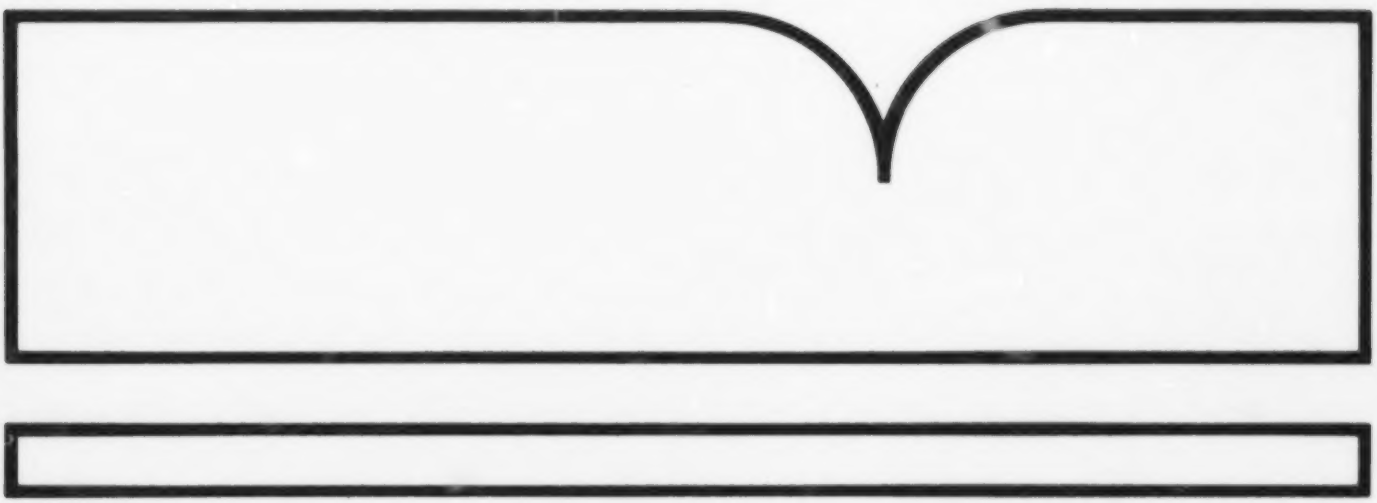

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by

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A hybrid receptor model is a specified mathematical procedure which uses not only the ambient species concentration measurements that form the input data for a pure receptor model, but in addition source emission rates or atmospheric dispersion or transformation information characteristic of dispersion models. By utilizing more information hybrid receptor modeling promises improved source apportionment estimates or, more fundamentally, consideration of problems that are inaccessible in terms of classical receptor modeling. Several examples of hybrid receptor models are reviewed, emphasizing the great variety in possible approaches, and in the choice of input versus output quantities. A simple illustration is given of a hybrid receptor model applied to the comprehensive ambient-source-meteorological data base collected at Deep Creek Lake, Maryland, during summer 1983.

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A hybrid receptor model is defined as a specified mathematical procedure which uses not only the ambient species concentration measurements that form the input data for a pure receptor model, but in addition source emission rates or atmospheric dispersion or transformation information characteristic of dispersion models. By utilizing more information hybrid receptor modeling promises improved source apportionment estimates or, more fundamentally, consideration of problems that are inaccessible in terms of classical receptor modeling. Several examples of hybrid receptor modeling are reviewed, emphasizing the great variety in possible approaches, and in the choice of input versus output quantities. A simple illustration is given of a hybrid receptor model applied to the comprehensive ambient-source-meteorological data base collected at Deep Creek Lake, Maryland during summer 1983.

A hybrid receptor model is any procedure for estimating the sources of ambient air pollutants at a receptor site, which makes use of both receptor and dispersion (source) modeling approaches. Thus, not only are the ambient species measurements which form the input data for a pure receptor model used, but also source emission rates or atmospheric dispersion or transformation information characteristic of dispersion

models. By exploiting simultaneously the strengths of the two complementary approaches we expect to minimize their individual weaknesses. The natural domain of hybrid receptor models is in the treatment of reactive air pollutants, such as particular organics, SO_2/sulfate and NO_x/nitrate. In the (non-exhaustive) review of existing hybrid receptor models which follows we hope to illustrate the variety of possible approaches, and the flexibility in the choice of input data versus calculated outputs, depending on what information is available. Finally, a comprehensive ambient-source-meteorological data base collected at Deep Creek Lake, Maryland, during summer 1983 is described and used in a simple application of one hybrid receptor model.

Hybrid Receptor Model Examples

Reactive Organic Chemical Mass Balance (Friedlander). In the original formulation of the CMB receptor model (1) it was recognized that the fractional amounts of various chemical species emitted by a source are not necessarily conserved during the transport of the species to the receptor site. This could occur through both physical (differential dispersion or deposition) or chemical (removal due to atmospheric reactions) processes. This possibility was acknowledged by writing the CMB equations in the form

$$C_i = \sum_j \alpha_{ij} a_{ij} S_j \qquad (1)$$

where C_i is the concentration of species i measured at the receptor site, a_{ij} (<1) is the mass fraction of species i in the emissions from source j, α_{ij} (<1) is the fractional decrease in a_{ij} during transport, and S_j is the (unknown) impact of source j at the receptor. Practically

every application of CMB, however, has set each decay factor α_{ij} to unity. An exception is the attempt by Friedlander (2) to derive an expression for decay factors to be used in the source apportionment of chemically reactive polycyclic aromatic hydrocarbons (PAH) in Los Angeles. Friedlander's result is

$$\alpha_{ij} = (1 + K_i\Theta)^{-1} \tag{2}$$

where K_i is the first order reaction rate constant for PAH species i, and Θ is its average residence time in the atmosphere. This result illustrates that a CMB receptor model that includes decay factors is actually a hybrid model, because the calculation of decay factor values requires information (reaction rates and dispersion considerations) that a strictly receptor approach cannot provide.

Tracer Hybrid Receptor Model (Lewis). Lewis and Stevens (3) have derived a hybrid receptor model for describing the secondary sulfate from an SO_2 point source. The resulting expression for secondary sulfate concentration M_{SO_4} at the receptor has the form

$$M_{SO_4} = M_p A T \tag{3}$$

with M_p being the mass concentration at the receptor of primary fine particles from the source (calculated from a classical receptor model), A representing the ratio of source mass emission rates for SO_2 and fine particles, and T describing both the transformation of SO_2 to sulfate in the atmosphere and its loss due to deposition. The function T has a simple analytical form depending on average values of the transformation and deposition rates, and the time of transit between source and receptor.

Thus, the calculation of secondary sulfate from Equation 3 can be thought of as the result of "grafting" additional emission ratio and wind speed information onto a conventional receptor modeling solution for the primary impact of the source. The principal advantage of Equation 3 is that all of the secondary sulfate dispersion -- and some of the deposition -- complexity is implicitly contained in M_p, which acts as the "tracer" for the source.

Gordon and Olmez (4) have used Equation 3 to model sulfate resulting from a multiple source distribution of coal-burning emissions, such as in the Ohio River Valley. Their results are quantitatively consistent with measured aerosol S/Se ratios of about 1700 and 3000, within and downwind of this region.

Source Finding Hybrid Receptor Model (Yamartino). The starting point of this model is a set of equations of the form

$$C_{kt} = \sum_j R_{jkt} Q_j \qquad (4)$$

where C_{kt} is the concentration of a pollutant at receptor site K during time interval t, Q_j is the emission rate of the pollutant from source j (an identifiable source or a source region), and R_{jkt} is the transfer coefficient between source j and receptor k for the meteorological conditions existent during time interval t. The C_{kt}'s are measured quantities, the R_{jkt}'s are to be calculated from a dispersion model (e.g., Gaussian plume), and the Q_j's are unknowns. The Q_j's are found by a least squares criteria, i.e., the correct set of Q_j's is assumed to be the one that minimizes

$$x^2 = \sum_{t,k} (C_{kt} - \sum_{j=1}^{J} R_{jkt}\, Q_j)^2 / (\Delta C_{kt})^2 \qquad (5)$$

where J is the number of source or source regions considered, and ΔC_{kt} is the uncertainty in C_{kt}. While Equation 4 bears a superficial resemblance to that of a conventional CMB receptor model approach, it is actually very different: the former is concerned with one pollutant observed at multiple receptors and times, while the latter involves multiple pollutants observed at one receptor and time; the source strengths in the former are those as observed at the source, while in the latter they describe the impact at the receptor; the former requires meteorological information (to calculate the R_{jkt}'s), while the latter has no such requirement.

Yamartino (5) has summarized the applications of this approach, which include determining the sources of CO at an airbase, the sources of SO_2 in West Berlin, and the wet deposition of sulfur in Eastern Northern America. The last application required estimates of rate constants as well as meteorological data to be inputted to calculate the transfer coefficients.

Diffusion Hybrid Receptor Model (Fay). This approach, beginning with the work of Fay and Rosenzweig (6), is perhaps the most interesting of all the hybrid models that have been proposed to date. Not only is it able to address the usual source apportionment problem of estimating source impacts (of SO_2 and secondary sulfate) at a receptor site but it simultaneously generates estimates for the conversion and deposition rate constants and meteorological parameters that are influencing the pollutant

transfer between source and receptor. Consequently, we choose to review this model in more detail than the others considered here.

The starting point of the model are Eulerian diffusion equations whose solutions are the ambient concentrations of primary (SO_2) sulfur, χ_p, and secondary (sulfate) sulfur, χ_s, resulting from a single SO_2 source having a known constant emission rate Q. It is important to appreciate that the diffusion equations are time-averaged, so that their proper application is the description of seasonal or annual concentration averages, certainly not daily ones. The solutions χ_p and χ_s, which are assumed to vary only with horizontal position, are zero order Bessel functions of the second kind. In addition to Q, χ_p and χ_s each depend on nine additional parameters: mixing height, wind speed and direction, horizontal diffusivity, first order rate constants for SO_2 conversion and wet and dry deposition, and first order rate constants for sulfate wet and dry deposition. All nine parameters are considered constant throughout the flow field.

Since the model is linear, the resultant prediction for the concentration of the primary or secondary pollutant at a receptor site is a linear superposition of the χ's for the individual point sources. The model is optimized by a least squares criteria: the nine parameter values are chosen so as to minimize E^2,

$$E^2 = \sum (\text{observation} - \text{prediction})^2 / \sum (\text{observation})^2 \qquad (6)$$

where the summation is over all observations.

The most successful application of this approach has been the source apportionment of the wet sulfate deposition rate in Eastern North America

(7). The parameter values resulting from the minimization of E^2 generally fall within the expected range for each one. The correlation coefficient between three year average predicted (using the optimized parameter set) wet deposition and observed values at 109 sites was 0.87. It is then a straightforward matter to calculate the percentage contribution from each source region at any chosen receptor site. For example, at Whiteface Mountain, New York, the three largest contributors to wet deposition were Ohio (11.9%), Pennsylvania (11.4%) and New York (7.8%). The apportionment of airborne sulfate was also accomplished using the same parameter values as obtained from the wet deposition problem.

The Deep Creek Lake Study

During August and September 1983 a field sampling study was conducted in rural Maryland at a site (Deep Creek Lake) which was anticipated to be heavily impacted by secondary sulfate from regional coal-fired power plants. When the analytical work in progress is completed (x-ray fluorescence, neutron activation analysis, ion chromatography), the resulting data base will consist of ambient aerosol measurements (130 six-hour intervals of size fractionated elemental and chemical concentration), ambient gas measurements (SO_2, O_3, and NO_x concentrations with 10 min time resolution), source measurements (size fractionated elemental aerosol composition and gaseous emission rates at 3 regional power plants expected to impact the receptor site), plume measurements (size fractionated aerosol composition at elevated points between sources and the receptor site), airborne aerosol regional signatures, and meteorologically-derived back trajectories. The unusually comprehensive content of this data set makes it a valuable resource for investigating various hybrid receptor model approaches.

Ambient Data Characteristics. Table I gives averages of selected fine particle species and gaseous sulfur measured at the Deep Creek Lake ambient site.

Table I. Fine Particle and Gaseous Sulfur Concentration Averages at Deep Creek Lake, MD, August 1983

Species	10 AM-10 PM (ng/m^3)	10 PM-10 AM (ng/m^3)
Mass	52700	39800
S_p*	8570	6630
$SO_4^=$	25700	19900
Pb	42.5	46.8
Se	4.9	4.9
S_g**	13900	15800
S_g/S_p (dimensionless)	1.8	3.3

*fine particle sulfur
**gaseous sulfur (from SO_2)

The table shows substantial fine mass loadings, with sulfate representing about half of the total. Selenium, which is increasingly regarded as a good tracer species for coal combustion, also has a high concentration relative to that found in airsheds with little coal impact. The concentration of lead, traditionally a good indicator of mobile source emissions, shows little impact by this source category.

Table II shows the linear correlation coefficients of the species from Table I. For 95 observations a correlation greater than 0.26 is significant at the 1% level. Thus, the correlations involving lead are not significant (consistent with expectation), while the mutual correlations of fine particle sulfur, selenium and mass are impressively large. *Correlations involving gaseous sulfur are significant, but less so.*

Table II. Linear Correlation Coefficients (N = 95)

	Mass	S_p	S_g	Se	Pb
S_p	.94				
S_g	.32	.32			
Se	.75	.73	.40		
Pb	.16	.16	.10	.03	
$S_p + S_g$	.50	.51	.98	.53	.12

Tracer Hybrid Receptor Model Application. Even though the Deep Creek Lake data base is not yet complete, in terms of all the chemical analyses that are anticipated to be included, it may be useful to give a simple illustration of how a hybrid receptor model can provide some insight into the data.

Table III shows ambient measurements obtained on August 5, 1983, between 10 AM and 4 PM. The concentrations are all well above their average daytime values. The S_g/S_p ratio is among the largest values seen during daytime sampling periods, indicating little gas to particle conversion has occurred, and suggestive of a short transport time. During this period back trajectory calculations showed an average wind speed of 14 km/h, from the direction of the nearest major coal-fired power plant (Ft. Martin, 1150 MW) 45 km due west of Deep Creek Lake. Table III also shows the measured S_g/Se ratio obtained at the Ft. Martin plant during September 1983. Dilution source sampling was employed so that the distribution of selenium between its fine particle and gaseous phases should approximate that which is present in the ambient environment.

Table III. Ambient and Source Fine Particle and Gaseous Sulfur Measurements

Deep Creek Lake, August 5, 1983, 10 AM - 4 PM		
S_g	49,300	ng/m^3 (37.6 ppb)
S_p	18,100	ng/m^3
Se	7.9	ng/m^3
S_g/S_p	2.7	
S_g/Se	6,240	
Ft. Martin Power Plant, September, 1983		
S_g/Se	21,000	

Under the assumption that the gaseous sulfur, fine particle selenium and secondary fine particle sulfur measured at an ambient site originates from a single point source the tracer hybrid receptor model can be expressed in terms of the two equations

$$S_p/S_g = [\exp((\overline{K}_r + \overline{K}_d)t)-1]\overline{K}_r t/(\overline{K}_r + \overline{K}_d)t \quad (7)$$

$$S_g/Se = (S_g/Se)_o \exp(-(\overline{K}_r + \overline{K}_d)t) \quad (8)$$

where t is the source to receptor transport time, $\overline{K}_r$ is the average gas to particle conversion rate, $\overline{K}_d$ is the average excess deposition rate for SO_2 relative to sulfate, and the zero subscript indicates a source measurement. Inserting the three ratios from Table III, Equations 7 and 8 have the unique solution (most easily obtained graphically) 0.19 and 1.0 for $\overline{K}_r t$ and $\overline{K}_d t$, respectively. Assuming the source to be the Ft. Martin plant, and thus a 3.2h transport time based on the back trajectory information, we have

$$\overline{K}_r = 5.9\ \%/h \quad (9)$$

$$\overline{K}_d = 31\ \%/h \quad (10)$$

The uncertainty in $\overline{K}_r$ is estimated to be 15%, while the uncertainty in $\overline{K}_d$ is at least 30%. The $\overline{K}_r$ value is similar to other measurements for dry summer daytime conditions. The $\overline{K}_r$ value is more difficult to compare, but is at least plausible.

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Literature Cited

1. Friedlander, S. Environ. Sci. Technol. 1973, 7, 235.

2. Friedlander, S. In Atmospheric Aerosol: Source/Air Quality Relationships; Macias, E. S.; Hopke, P. K., Eds.; ACS Symposium Series No. 167; American Chemical Society: Washington, DC, 1981; p. 1.

3. Lewis, C. W., Stevens, R. K. Atmos. Environ. 1985, 19, 917.

4. Gordon, G. E.; Olmez, I. In Receptor Methods for Source Apportionment; Pace, T. G., Ed.; APCA International Specialty Conference; Air Pollution Control Association: Pittsburgh, PA, 1986; p. 229.

5. Yamartino, R. J. Proc. 77th Ann. Mtg. Air Pollu. Control Assoc., 1984, Paper No. 84-48.2.

(6) Fay, J. A.; Rosenzweig, J. J. Atmos. Environ. 1980, 14, 355.

(7) Fay, J. A.; Golomb, D.; Kumar, S. In Receptor Methods for Source Apportionment; Pace, T. G., Ed.; APCA International Specialty Conference; Air Pollution Control Association: Pittsburgh, PA, 1986; p. 105.